

This Page Is Inserted by IFW Operations
and is not a part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representation of
The original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

IMAGES ARE BEST AVAILABLE COPY.

**As rescanning documents *will not* correct images,
please do not report the images to the
Image Problem Mailbox.**

THIS PAGE BLANK (USPTO)

IE 970280



**Oifig Na
bPaitinní**

**The
Patents
Office**

ABSTRACT

Plastics material and a method for its manufacture

In an improved method of manufacturing biodegradable plastics material, a polyvinylalcohol and polyvinylalcohol/polyvinylacetate co-polymer (PVA/PVAc) are mixed with a plasticiser and stabiliser under conditions of applied physical force and temperature which enable the co-polymer to be worked without any significant degradation. The stabiliser may be any surface active agent, dispersing agent and/or mould lubricant, except steramide or stearic acid when used at a mixing temperature of between 106 and 140°C. A blend of stabilisers may be used. The stabiliser enhances the action of the plasticiser and maintains the stability of the resultant compound enabling it to be reproducibly thermoplastically processed and to yield products with minimal discoloration.

**TRUE COPY
AS
LODGED****IE 970280**

- 1 -

No.

PLASTICS MATERIAL AND A METHOD FOR ITS MANUFACTURE

The present invention relates to a biodegradable plastic compound for use in thermoplastic applications and its method of manufacture, and in particular to the biodegradable materials comprising polyvinylalcohol (PVA) and polyvinylalcohol/polyvinylacetate co-polymer, (PVA/PVAc).

PVA and PVA/PVAc co-polymers are known biodegradable materials. PVA and PVA/PVAc (of greater than 85% PVA) are soluble in warm and hot water. PVA/PVAc (of less than 85% PVA content) is readily soluble in water at room temperature. Post dissolution these materials will readily bio-degrade on exposure to bacteria present in the environment. These polymers, part degradation products and degradation products are non-toxic and pose no risk of bio-accumulation or long term hazard to the environment.

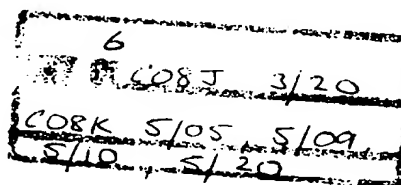
The applications of PVA and PVA/PVAc in thermoplastics have been limited by degradation which can occur on thermal processing.

German Patent No. 1127085 discloses that in order to process PVA/PVAc for thermoplastic applications, eg blown film moulding, extrusion or injection moulding, undesirable decomposition occurs at temperatures above 150°C and must be prevented by plasticising the granular plastics material. The plastics material is plasticised by mixing a suitable plasticiser for 15-30 minutes at a temperature of 110°C. The plasticised PVA/PVAc can then be injection moulded and extruded under the following conditions; heating the material to a temperature in the range of 149-190°C and at an injection pressure of 100-130 bar.

Irish Patent No. S71912 discloses that PVA/PVAc co-polymer comprising 70-85% PVA and 30-15% PVA with molecular weights in the range 20,000 to 90,000 can be more efficiently plasticised by the addition of 3-15% weight equivalent glycerol with the aid of 4-6% weight equivalent of stearamide or stearic acid salts as a 'stabiliser'. The mixing of

**OPEN TO PUBLIC INSPECTION
UNDER
SECTION 28 AND RULE 23**

JNL No. 1821 OF 24/9/97



components is carried out at 106-140°C and the material processed at 195-225°C into thermoplastic materials.

Variable levels of thermal degradation can occur to PVA/PVAc co-polymer, even when
5 plasticised, depending upon the energy applied to the copolymer in the form of temperature, time exposed to temperature and mechanical energy applied during processing. The variable and erratic nature of these factors in conventional manufacturing have rendered these materials unpredictable in nature and difficult to use. Also, excessive degradation would alter the physical properties of the plastics material, rendering it
10 mechanically weaker (than non-degraded material) and discoloured, and potentially alter its bio-degradation properties.

Unplasticised PVA undergoes significant decomposition on melting rendering it impractical as a thermoplastic. Even, plasticised PVA/PVAc may decompose by more
15 than 5% when conventionally processed above 200°C. The degree of decomposition can be reduced to insignificant levels working with plasticised and stabilised PVA/PVAc below 185°C

The present invention seeks to provide an improved method of manufacturing an article or
20 material using PVA/PVAc.

Accordingly, the present invention provides a method of manufacturing a biodegradable plastics material, comprising mixing a polyvinylalcohol polymer or polyvinylalcohol/polyvinylacetate copolymer with a plasticiser or a blend of plasticisers
25 and a stabiliser or blend of stabilisers, the stabiliser or blend thereof comprising a surface active agent, dispersing and/or mould lubricant except stearamide or stearic acid salts when used as single stabiliser and at a mixing temperature in the range of between 106 and 140°C, the plasticiser and stabiliser or blends thereof rendering the resultant polyvinylalcohol polymer or polyvinylalcohol/polyvinylacetate copolymer compound
30 stable to degradation by thermoplastics processing. Conveniently, a blend of stabilisers is used to enhance the effectiveness of the plasticiser by rendering the molten plastic more

- 3 -

fluid and easier to process than can be achieved using a single stabiliser. The plasticiser may be selected from any low-volatile alcohol, ester or ether, or any bi- or tri-functional alcohol, ester or ether or any combination thereof. The plasticiser preferably comprises any material selected from :-

5

(1) Glycerol, propylene glycol, 1,3-propanediol, ethylene glycol; or

(2) any mono-, di- or tri- (C1 to C10) carboxylic acid esters of materials listed (1) above; or

10

(3) citric acid (C1 to C4) alcohol tri-esters or mixed tri-esters or glycol esters; or

(4) tartaric acid (C1 to C4) alcohol di-esters or mixed di-esters or glycol esters; or

15

(5) lactic acid (C1 to C4) alcohol esters, ethylene or propylene glycol esters; or

(6) malic acid (C1 to C4) alcohol di-esters or mixed di-esters or glycol esters; or

(7) glyceric acid (C1 to C4) alcohol esters or ethylene glycol ester; or

20

(8) any (C1 to C4) ether, glycol ether or mixed ether of materials listed (1) to (7) above; or

(9) any (C1 to C4) acetal or ketal derivative of materials listed (1) to (7) above; or

25

(10) any vegetable base oils including soya oil or corn oil; or

(11) any (C1 to C10) mono-, di- or tri- ester of adipic acid, azelaic acid, phthalic acid or trimellitic acid;

30

or any combination of (1) to (11) above

and the stabiliser comprises any material selected from:-

(12) any (C4 to C36) mono- or di-carboxylic acid; or

5

(13) any primary, secondary or tertiary amide or diamide of (12) above; or

(14) any inorganic or organic salt of (12) above; or

10

(15) any mono-, di- or tri glycerol ester or mixed glycerol ester of (12) above; or

(16) any ethylene glycol, propylene glycol or 1,3- propanediol ester of (12) above;
or

15

(17) any gum resin materials used as dispersing aids; or

(18) any wax ionomer or dispersing aid; or

(19) any sorbitan fatty ester; or

20

(20) any sorbitan polyoxyethylene ester; or

(21) any polyoxyethylene glycol (C1 to C30) ester; or

25

(22) any polymeric based hyper-dispersant; or

any combination of (12) to (22) above.

In a preferred method, the plasticiser includes triacetin and the stabiliser includes glycerol
30 mono- stearate. The stabiliser may also include a blend of stabilisers including a stearic
acid salt and/or stearamide.

Conveniently, the mixing is carried out at a temperature of at least 55°C to form a plasticised compound which can be thermoplastically processed at a temperature of at least 120°C.

5

The plasticiser or blend thereof is preferably used in an amount from 2-30% weight equivalents of the polyvinylalcohol polymer or polyvinylalcohol/polyvinylacetate copolymer. The stabiliser or blends thereof is preferably used in an amount from 2-6% weight equivalents of the polyvinylalcohol polymer or polyvinylalcohol/polyvinylacetate copolymer.

10

The invention also provides a biodegradable article or material manufactured from a plastics compound prepared from a mixture of a polyvinylalcohol polymer or polyvinylalcohol/polyvinylacetate copolymer, a plasticiser or a blend of plasticiser and a stabiliser or a blend of stabiliser, the stabiliser or blend thereof comprising a surface active agent, dispersing and/or mould lubricant except stearamide or stearic acid salts when used as sole stabiliser and at a mixing temperature in the range of between 106 and 140°C, the plasticiser and stabiliser or blends thereof rendering the plastics compound stable to degradation by thermoplastic processing used in the manufacture of an article or material from the plastics compound.

15

20

The present invention provides an improved method for the manufacture of biodegradable plastic materials comprising mixing PVA/PVAc co-polymer (70-95% PVA and 30-5% PVAc) with systems of 'plasticiser' and 'stabiliser' at temperatures from 55-150°C. Mixing is typically carried out in a force action blender or other conventional polymer mixing apparatus. The exact mixing temperatures reached have been found to be non-critical. The key parameters to efficiency of the plasticising process have been found to be mechanical efficiency, temperature and time in combination. Thus, high speed mixing of a compound previously made at a high temperature can be carried out at lower temperature with less vigorous mixing but would require a longer cycle time.

25

30

The resultant material can be conventionally processed in numerous thermoplastic applications at temperatures of 120-225°C, eg; into blown film, sheet or other extrusion, injection moulded articles, woven or non-woven fibres or expanded foam products, or compounded as pellets for re-use in the aforementioned applications.

5

The action attributed to the stabiliser is to enhance the action of the plasticiser and maintain the stability of the resultant compound. The invention relates to the use of combinations of materials in the stabiliser system, which tends to magnify the system's effectiveness.

- 10 The materials produced by the present invention are capable of being manipulated at lower temperatures than previously disclosed, thereby incurring minimal thermal degradation. Therefore, the materials produced can be worked and re-worked with greater degree of control and reproducibility of physical performance in application and much less visible discoloring of the material.

15

- The present invention also enables greater fluidity of the melted plastic material so that the material may be processed at lower temperature and/or pressure, by the use of comparably lower amounts of stabiliser than previous disclosures (2-6% weight equivalents), thereby achieving wider capability of thermoplastics application at potentially lower cost. For
20 example, the present invention renders the material capable of being worked at comparatively low pressures and temperatures of 40-75 bar (kN/m²) at 120-185°C, whereas the invention described in Irish Patent No. S71912 discloses typical pressures of 40-75 bar (kN/m²) at temperatures 195-225°C.

- 25 The plasticiser employed may be used in the range 2-30% weight equivalents. Plasticiser levels of 2-15% are generally preferred and the most preferred plasticisers are low-volatile liquids.

- As used herein, the term "low-volatile" refers to any material which is normally liquid at
30 20°C and 1 bar (1 MPa) and whose boiling point is greater than or equal to 150°C at 1 bar (1 MPa).

The above processes have the capability to produce a range of compounded plastic materials with varying physical characteristics, eg; melt flow index, strength, flexibility and rate of dissolution, depending upon the degree of hydrolysis of the PVA/PVAc copolymer, its average molecular weight and the qualities and type of Plasticiser(s) and Stabiliser(s) employed. The present invention includes the manufacture of such a range and its use to provide a plastics processor with materials which can be used solely or in any combination so that he may obtain materials to match his specific desired product performance criteria suitable for the manufacture of a wide variety of plastic articles with different performance needs.

Materials such as those whose manufacture is described in Examples 5 (strong but moderate melt flow index insufficient to fill complex moulding) and 9A (weaker but high melt flow index) herebelow may be blended to give a material which is both strong and of high melt flow index providing an optimum in performance between the two materials.

PVA/PVAc copolymer films are known to exhibit greater strength than corresponding low density polyethylene films. Where an article of packaging is manufactured from PVA/PVAc, it may offer too much resistance to being torn open by the consumer and hence represent an inconvenience in opening. The present invention allows materials with a wide range of physical performance to be manufactured, making possible the production of films which are strong in application but may be torn open with relative ease by a consumer.

The 'Plasticiser' may be any material indicated in Schedule A or any combination of materials indicated in Schedule A.

The 'Stabiliser' may be any compound listed in Schedule B. (excluding stearamide or stearic acid salts when used on their own as sole stabiliser and the compound mixed at a temperature in the range of 106-140°C), or any combination of materials listed in Schedule B (including stearamide and stearic acid salts).

The materials listed in Schedules A and B hereunder are not limiting and other suitable materials or combinations of materials will suggest themselves to the skilled person.

- 5 The invention will now be described more particularly with reference to the following examples.

Example 1

Ingredients:

- 10 PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 50,000 - 100 parts

Glycerol - 15 parts

- The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 125-135°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC
15 twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die. Temperatures at the feed-zone, work-zone and die zone were; 170°C, 195°C and 170°C respectively. Working pressures were 60-95 bar (6-9.5 MPa). Pale yellow Pellet and film were obtained.

20

Example 2

Ingredients:

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 50,000 - 100 parts

Glycerol - 10 parts

- 25 The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 125-135°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die. Temperatures at the feed-zone, work-zone and die zone were; 170°C, 200°C and 195°C
30 respectively. Working pressures were 60-95 bar (6-9.5 MPa). Pale yellow Pellet and film were obtained.

Example 3

Ingredients:

5 PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 50,000 - 100 parts

Glycerol - 5 parts

The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 125-135°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC
10 twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die. Temperatures at the feed-zone, work-zone and die zone were: 170°C, 210°C and 210°C respectively. Working pressures were 60-95 bar (6-9.5 MPa). Pale yellow Pellet and film were obtained.

15

Example 4

Ingredients:

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 50,000 - 100 parts

Glycerol - 10 parts

20 Stearamide - 5 parts

The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 120-135°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die.
25 Temperatures at the feed-zone, work-zone and die zone were: 160°C, 195°C and 170°C respectively. Working pressures were 60-95 bar (6-9.5 MPa). Good quality Pellet and film were obtained.

30 Example 5

Ingredients:

- 10 -

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 50,000 - 100 parts

Glycerol - 15 parts

Stearamide - 2 parts

Calcium Stearate - 1 part

- 5 The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 80-135°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die. Temperatures at the feed-zone, work-zone and die zone were; 160°C, 183°C and 140°C
10 respectively. Working pressures were 40-75 bar (4-7.5 MPa). Good quality Pellet and film were obtained.

Example 6

- 15 Ingredients:

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 50,000 - 100 parts

Glycerol - 10 parts

Stearamide - 2.6 parts

Calcium Stearate - 1.3 parts

- 20 The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 80-135°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die. Temperatures at the feed-zone, work-zone and die zone were; 160°C, 195°C and 155°C
25 respectively. Working pressures were 50-85 bar (5-8.5 MPa). Good quality Pellet and film were obtained.

Example 7

- 30 Ingredients:

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 50,000 - 100 parts

Glycerol - 5 parts

Stearamide - 3.33 parts

Calcium Stearate - 1.66 parts

The Ingredients were combined and mixed in a force action blender between 1,500 and
5 3,000 rpm, until a temperature of 80-135°C was reached. The resultant powder was cooled
to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin
screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die.
Temperatures at the feed-zone, work-zone and die zone were; 160°C, 195°C and 170°C
respectively. Working pressures were 50-85 bar (5-8.5 MPa). Good quality Pellet and
10 film were obtained.

Example 8A

Ingredients:

15 PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 80,000 - 100 parts

Glycerol - 15 parts

Stearamide - 2 parts

Calcium Stearate - 1 part

The Ingredients were combined and mixed in a force action blender between 1,500 and
20 3,000 rpm, until a temperature of 80-135°C was reached. The resultant powder was cooled
to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin
screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die.
Temperatures at the feed-zone, work-zone and die zone were; 160°C, 187°C and 159°C
respectively. Working pressures were 65-95 bar (6.5-9.5 MPa).
25 Good quality Pellet and film were obtained.

Example 8B

Ingredients:

30 PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 80,000 - 100 parts

Glycerol - 15 parts

Stearamide - 4 parts

Calcium Stearate - 2 part

The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 80-135°C was reached. The resultant powder was cooled
5 to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die. Temperatures at the feed-zone, work-zone and die zone were; 153 °C, 185°C and 153°C respectively. Working pressures were 55-90 bar (5.5-9 MPa). Good quality Pellet and film were obtained.

10

Example 9A

Ingredients:

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 20,000 - 100 parts

15 Glycerol - 15 parts

Stearamide - 2 parts

Calcium Stearate - 1 part

The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 80-123°C was reached. The resultant powder was cooled
20 to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die. Temperatures at the feed-zone, work-zone and die zone were; 150°C, 170°C and 120 °C respectively. Working pressures were 40-75 bar (4-7.5 MPa). Good quality Pellet and film were obtained.

25

Example 9B

Ingredients:

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 20,000 - 100 parts

30 Glycerol - 15 parts

Stearamide - 2.7 parts

Calcium Stearate - 1.3 part

The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 80-123°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die. Temperatures at the feed-zone, work-zone and die zone were; 130°C, 174°C and 129°C respectively. Working pressures were 50-85 bar (5-8.5 MPa). Good quality Pellet and film were obtained.

10

Example 9C

Ingredients:

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 20,000 - 100 parts

Glycerol - 15 parts

15 Stearamide - 3.3 parts

Calcium Stearate - 1.7 part

The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 80-123°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die. Temperatures at the feed-zone, work-zone and die zone were; 140°C, 172°C and 137°C respectively. Working pressures were 50-85 bar (5-8.5 MPa). Good quality Pellet and film were obtained.

25

Example 9D

Ingredients:

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 20,000 - 100 parts

Glycerol - 15 parts

30 Stearamide - 1.3 parts

Calcium Stearate - 0.7 part

The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 80-123°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die.

5 Temperatures at the feed-zone, work-zone and die zone were; 150°C, 170°C and 125°C respectively. Working pressures were 50-85 bar (5-8.5 MPa). Good quality Pellet and film were obtained.

10 Example 9E

Ingredients:

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 20,000 - 100 parts

Glycerol - 2 parts

Stearamide - 2 parts

15 Calcium Stearate - 1 part

The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 80-123°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die.

20 Temperatures at the feed-zone, work-zone and die zone were; 100 °C, 158°C and 144°C respectively. Working pressures were 45-65 bar (4.5-6.5 MPa). Good quality Pellet and film were obtained.

25 Example 10

Ingredients:

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 50,000 - 100 parts

Glycerol - 15 parts

Glycerol mono-stearate - 2 parts

30 Calcium Stearate - 1 part

- 15 -

The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 80-123°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die.

5 Temperatures at the feed-zone, work-zone and die zone were; 152°C, 178°C and 136°C respectively. Working pressures were 60-80 bar (6-8 MPa). Good quality Pellet and film were obtained.

10 Example 11A

Ingredients:

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 50,000 - 100 parts

Glycerol - 12 parts

Triacetin - 3 parts

15 Stearamide - 2 parts

Calcium Stearate - 1 part

The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 80-123 °C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC

20 twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die. Temperatures at the feed-zone, work-zone and die zone were; 152 °C, 175°C and 129°C respectively. Working pressures were 60-80 bar (6-8 MPa). Good quality Pellet and film were obtained.

25

Example 11B

Ingredients:

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 50,000 - 100 parts

Glycerol - 12 parts

30 Dioctyl adipate - 3 parts

Stearamide - 2 parts

Calcium Stearate - 1 part

The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 80-123°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin
5 screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die. Temperatures at the feed-zone, work-zone and die zone were: 152°C, 183°C and 134°C respectively. Working pressures were 60-80 bar (6-8 MPa). Good quality Pellet and film were obtained.

10

Example 11C

Ingredients:

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 50,000 - 100 parts

Triacetin - 15 parts

15

Stearamide - 2 parts

Calcium Stearate - 1 part

The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 80-123°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin
20 screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die. Temperatures at the feed-zone, work-zone and die zone were: 155°C, 174°C and 160°C respectively. Working pressures were 60-80 bar (6-8 MPa). Good quality Pellet and film were obtained.

25

Example 11D

Ingredients:

PVA/PVAc (75-80%PVA:25-20%PVAc), Ave. MWt 50,000 - 100 parts

Triacetin - 15 parts

30

Glycerol mono-stearate - 2 parts

Calcium Stearate - 1 part

- 17 -

The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 80-123°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die.

5 Temperatures at the feed-zone, work-zone and die zone were; 155°C, 174°C and 160°C respectively. Working pressures were 60-80 bar (6-8 MPa). Good quality Pellet and film were obtained.

10 Example 12

Ingredients:

PVA/PVAc (80-95%PVA:5-10%PVAc), Ave. MWt 40,000 - 100 parts

Glycerol - 10 parts

Stearamide - 2 parts

15 The Ingredients were combined and mixed in a force action blender between 1,500 and 3,000 rpm, until a temperature of 120-135°C was reached. The resultant powder was cooled to room temperature, sieved and compounded as pellet using a Prism TSE 16TC twin screw extruder with a 4.5mm die or Blown as film using 90°, 30 mm Blow die. Temperatures at the feed-zone, work-zone and die zone were; 170°C, 195 °C and 185°C

20 respectively. Working pressures were 65-95 bar (6.5-9.5 MPa). Slightly pale Pellet and film were obtained.

SCHEDULE A

- 25 (1) Glycerol, propylene glycol, 1,3-propanediol, ethylene glycol.
- (2) Any mono-, di- or tri- (C1 to C10) carboxylic acid esters of materials listed (A1) above.
- 30 (3) Citric acid (C1 to C4) alcohol tri-esters or mixed tri-esters or glycol esters.

- 18 -

- 5 (4) Tartaric acid (C1 to C4) alcohol di-esters or mixed di-esters or glycol esters.
- (5) Lactic acid (C1 to C4) alcohol esters, ethylene or propylene glycol esters.
- 5 (6) Malic acid (C1 to C4) alcohol di-esters or mixed di-esters or glycol esters.
- (7) Glyceric acid (C1 to C4) alcohol esters or ethylene glycol ester.
- 10 (8) Any (C1 to C4) ether, glycol ether or mixed ether of materials listed (A1) to (A7) above.
- (9) Any (C1 to C4) acetal or ketal derivative of materials listed (A1) to (A7) above.
- 15 (10) Any vegetable base oils eg.; soya oil or corn oil.
- (11) Any (C1 to C10) mono-, di- or tri- ester of adipic acid, azelaic acid, phthalic acid or trimellitic acid.
- 20 (12) Any combination of the above.

SCHEDULE B

- 25 (1) Any (C4 to C36) mono- or di-carboxylic acid
- (2) Any primary, secondary or tertiary amide or diamide of (B1) above.
- (3) Any inorganic or organic salt of (B1) above.
- 30 (4) Any mono-, di- or tri glycerol ester or mixed glycerol ester of (B1) above.

- 5
- (5) Any ethylene glycol, propylene glycol or 1,3- propanediol ester of (B1) above
- (6) Any gum resin materials used as dispersing aids, eg. Permalyn(R) 5095 or 5110 (Trademark; Hercules Chemicals).
- (7) Any wax ionomer or dispersing aid; eg. Aclyn(R) 295A (Trademark; Allied Signal Inc).
- 10 (8) Any sorbitan fatty ester; eg. Span(R) 20 to 85, (Trademark; Atlas Chemical Inc.) etc.
- (9) Any sorbitan polyoxyethylene ester; eg Tween(R) 20 to 85, (Trademark; Atlas Chemical Inc.) etc.
- 15 (10) Any polyoxyethylene glycol (C1 to C30) ester; eg Brij(R) 30 to 96 (Trademark of Atlas Powder), etc.
- (11) Any polymeric based hyper-dispersant; eg Solsperse(R) 20000 or 27000 (Trademark of Zeneca), etc.
- 20 (12) Any combination of the above.

It will of course be understood that the invention is not limited to the specific details described herein, which are given by way of example only, and that various modifications and alterations are possible within the scope of the invention as defined in the appended claims.

25

CLAIMS:

1. A method of manufacturing a biodegradable plastics material, comprising mixing a polyvinylalcohol polymer or polyvinylalcohol/polyvinylacetate copolymer with a plasticiser or a blend of plasticisers and a stabiliser or blend of stabilisers, the stabiliser or blend thereof comprising a surface active agent, dispersing and/or mould lubricant except stearamide or stearic acid salts when used as single stabiliser and at a mixing temperature in the range of between 106 and 140°C, the plasticiser and stabiliser or blends thereof rendering the resultant polyvinylalcohol polymer or polyvinylalcohol/polyvinylacetate copolymer compound stable to degradation by thermoplastics processing.

2. A method according to Claim 1, in which a blend of stabilisers is used to enhance the effectiveness of the plasticiser by rendering the molten plastic more fluid and easier to process than can be achieved using a single stabiliser and the plasticiser is selected from any low-volatile alcohol, ester or ether, or any bi- or tri-functional alcohol, ester or ether or any combination thereof,

optionally wherein the plasticiser comprises any material selected from :-

- (1) Glycerol, propylene glycol, 1,3-propanediol, ethylene glycol; or
- (2) any mono-, di- or tri- (C1 to C10) carboxylic acid esters of materials listed (1) above; or
- (3) citric acid (C1 to C4) alcohol tri-esters or mixed tri-esters or glycol esters; or
- (4) tartaric acid (C1 to C4) alcohol di-esters or mixed di-esters or glycol esters; or
- (5) lactic acid (C1 to C4) alcohol esters, ethylene or propylene glycol esters; or
- (6) malic acid (C1 to C4) alcohol di-esters or mixed di-esters or glycol esters; or

(7) glyceric acid (C1 to C4) alcohol esters or ethylene glycol ester; or

5 (8) any (C1 to C4) ether, glycol ether or mixed ether of materials listed (1) to (7) above; or

(9) any (C1 to C4) acetal or ketal derivative of materials listed (1) to (7) above; or

10 (10) any vegetable base oils including soya oil or corn oil; or

(11) any (C1 to C10) mono-, di- or tri- ester of adipic acid, azelaic acid, phthalic acid or trimellitic acid;

15 or any combination of (1) to (11) above

and the stabiliser comprises any material selected from:-

(12) any (C4 to C36) mono- or di-carboxylic acid; or

20 (13) any primary, secondary or tertiary amide or diamide of (12) above; or

(14) any inorganic or organic salt of (12) above; or

25 (15) any mono-, di- or tri glycerol ester or mixed glycerol ester of (12) above; or

(16) any ethylene glycol, propylene glycol or 1,3- propanediol ester of (12) above; or

30 (17) any gum resin materials used as dispersing aids; or

(18) any wax ionomer or dispersing aid; or

(19) any sorbitan fatty ester; or

(20) any sorbitan polyoxyethylene ester; or

5

(21) any polyoxyethylene glycol (C1 to C30) ester; or

(22) any polymeric based hyper-dispersant; or

10 any combination of (12) to (22) above,

optionally in which the plasticiser includes triacetin.

optionally in which the stabiliser includes glycerol mono- stearate,

15

optionally in which the stabiliser comprises a blend of stabilisers including a stearic acid salt and/or stearamide,

optionally in which the mixing is carried out at a temperature of at least 55°C to form a
20 plasticised compound,

optionally in which the plasticised compound can be thermoplastically processed at a temperature of at least 120°C,

25 optionally in which the plasticiser or blend thereof is used in an amount from 2-30% weight equivalents of the polyvinylalcohol polymer or polyvinylalcohol/polyvinylacetate copolymer, and

optionally in which the stabiliser or blends thereof is used in an amount from 2-6% weight
30 equivalents of the polyvinylalcohol polymer or polyvinylalcohol/polyvinylacetate copolymer.

3. A biodegradable article or material manufactured by a method according to either Claims 1 or 2.
- 5 4. A biodegradable article or material manufactured from a plastics compound prepared from a mixture of a polyvinylalcohol polymer or polyvinylalcohol/polyvinylacetate copolymer, a plasticiser or a blend of plasticiser and a stabiliser or a blend of stabiliser, the stabiliser or blend thereof comprising a surface active agent, dispersing and/or mould lubricant except stearamide or stearic acid salts when used as sole stabiliser and at a mixing
10 temperature in the range of between 106 and 140°C, the plasticiser and stabiliser or blends thereof rendering the plastics compound stable to degradation by thermoplastic processing used in the manufacture of an article or material from the plastics compound.
- 15 5. A biodegradable article or material or a method of manufacturing a biodegradable plastics article or material, substantially as herein described with reference to the Examples.

20

MACLACHLAN & DONALDSON,
Applicants' Agents,
47 Merrion Square,
DUBLIN 2.